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(54) **[Title of the Invention] Method for Manufacturing Polyphenylene Ether Tablets**

(57) [Abstract]

[Object] A method for manufacturing polyphenylene ether that is easy to handle, is very safe, and has good extruder bite.

[Constitution] A method for manufacturing polyphenylene ether, in which a polyphenylene ether powder that has been heated to a temperature between 70 and 200°C is subjected to compression molding at a pressure between 0.1 and 5.0 tons/cm^2, which yields solid tablets with a density between 0.7 and 1.5.

[Claims]

[Claim 1] A method for manufacturing polyphenylene ether, wherein a polyphenylene ether powder that has been heated to a temperature between 70 and 200°C is subjected to compression molding at a pressure between 0.1 and 5.0 tons/cm^2, which yields solid tablets with a density between 0.7 and 1.5.

[Claim 2] A method for manufacturing polyphenylene ether as defined in Claim 1, wherein the size of the solid tablets is between 0.1 and 10 mm.

Detailed Description of the Invention

[0001]

Technological Field to Which the Invention Belongs

The present invention relates to a method for manufacturing tablets of a polyphenylene ether resin that is easier to handle in the form of a powder.

[0002]

Prior Art

Techniques for manufacturing a polyphenylene ether by subjecting a phenolic compound to oxidation polymerization have been discussed in Japanese Patent Publication S42-3195, Japanese Laid-Open Patent Application S52-897, and elsewhere. The method employed to recover a polyphenylene ether resin from the polymer solution produced with one of these manufacturing methods generally involved precipitating and recovering the polymer by the addition of methanol or another such poor solvent of polyphenylene ether. The polymer particles precipitated here included both extremely fine particles and large particles. Accordingly, the powder was difficult to handle, tending to scatter, and furthermore, considerable care had to be taken, in terms of safety, to prevent dust explosion or the like. Also, because of the poor powder characteristics, insufficient bite during extrusion granulation of the resin, and so forth, these methods resulted in inferior productivity.

[0003] Various methods have been tried in an effort to solve these problems. In Japanese Laid-Open Patent Application H7-97441 is discussed an attempt to reduce scattering by adding a surfactant to the powder particles so as to eliminate the static charge generated in the powder.

[0004]

Problems Which the Invention is Intended to Solve

For the effect they provided, however, these methods were complicated and therefore more costly. There has been a need for a way to make a polyphenylene ether powder easier to handle with a simpler and more efficient method.

[0005]

Means Used to Solve the Above-Mentioned Problems

As a result of diligent research conducted in order to solve the above problems, the inventors arrived at the present invention upon discovering that solid tablets of adequate strength can be easily obtained by applying a specific pressure to a polyphenylene ether powder that has been heated to a specific temperature.

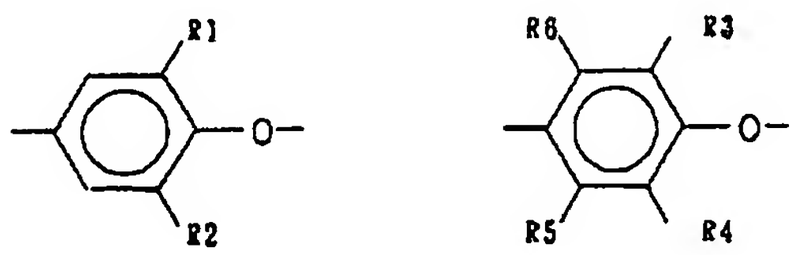
[0006] Specifically, the present invention is a manufacturing method in which a polyphenylene ether powder that has been heated to a temperature between 70 and 200°C is subjected to compression molding at a pressure between 0.1 and 5.0 tons/cm^2, which yields solid tablets with a density between 0.7 and 1.5.

[0007] The present invention will now be described in further detail.

[0008] The polyphenylene ether resin used in the present invention is a homopolymer or copolymer having repeating units expressed by the following general formula (1) and having structural units composed of the following [a] and [b].

[0009]

[First Chemical Formula]



(1)

[a] [b]

[0010] Typical examples of a homopolymer of a polyphenylene ether-based resin include poly(2,6-dimethyl-1,4-phenylene) ether, poly(2-methyl-6-ethyl-1,4-phenylene) ether, poly(2,6-diethyl-1,4-phenylene) ether, poly(2-ethyl-6-n-propyl-1,4-phenylene) ether, poly(2,6-di-n-propyl-1,4-phenylene) ether, poly(2-methyl-6-n-butyl-1,4-phenylene) ether, poly(2-ethyl-6-isopropyl-1,4-phenylene) ether, poly(2-methyl-6-chloroethyl-1,4-phenylene) ether, poly(2-methyl-6-hydroxyethyl-1,4-phenylene) ether, and other such homopolymers.

[0011] Polyphenylene ether copolymers encompass a copolymer of 2,6-dimethylphenol and 2,3,6-trimethylphenol, a copolymer of [2,6-dimethylphenol and] o-cresol, a copolymer of 2,3,6-trimethylphenol and o-cresol, and other such polyphenylene ether copolymers whose main component is a polyphenylene ether structure.

[0012] Also, to the extent that the object of the present invention is not compromised, the polyphenylene ether-based resin of the present invention may also include as part of its structure any of various other phenylene ether units, which it has been proposed in the past may be present in a polyphenylene ether resin. Examples of those which have been proposed may be present in a small amount include 2-(dialkylaminomethyl)-6-

methylphenylene ether units and 2-(N-alkyl-N-phenylaminomethyl)-6-methylphenylene units, which are discussed in Japanese Laid-Open Patent Applications S63-12698 and S63-301222.

[0013] A small amount of diphenoquinone or the like may also be bonded to the main chain of the polyphenylene ether.

[0014] Also included are polyphenylene ethers modified by compounds having carbon-carbon double bonds, which are discussed in Japanese Laid-Open Patent Applications H2-276823, S63-108059, and S59-59724.

[0015] The molecular weight of the polyphenylene ether resin used in the present invention is 1000 to 100,000, as the number average molecular weight. A preferred range thereof is 6000 to 60,000. The "number average molecular weight" referred to in the present invention is the number average molecular weight calculated for polystyrene as determined using a standard polystyrene calibration curve by gel permeation chromatography.

[0016] Examples of the compression apparatus used in the present invention include a compression roll type, pre-getting roll type, tablet-molding type, and other such compression molding machines, but the effect of the present invention will be thoroughly achieved as long as a pressure of 0.1 to 5.0 tons/cm^2 can be applied in a 70 to 200°C heated state.

[0017] It is conceivable that the size of the molded article might be over 10 mm with some molding machines, but the effect of the present invention can still be obtained by molding an article of 10 mm or larger by the process of the present invention and then breaking it up into pieces 0.1 to 10 mm in size.

[0018] With a conventional method, the compressed product did not have enough compression strength for it to withstand practical use, but the use of the manufacturing conditions of the present invention makes it possible to obtain tablets with adequate compression strength.

[0019] The temperature range of the powder during manufacture must be between 70 and 200°C. Tablets of adequate strength will not be obtained below 70°C, but if 200°C is exceeded the amine contained in the polyphenylene ether will be dissociated and the quality [of the product] will be decreased during working. Preferably, the temperature should be between 100 and 150°C, in which case it will be possible to obtain substantially stable tablets that are more durable and have better surfaces. The pressure during manufacture must be between 0.1 and 5.0 tons/cm^2. Tablets of adequate strength will not be obtained below 0.1 ton/cm^2, but if 5.0 tons/cm^2 is exceeded, the tablets will fuse, the density will rise, and it will be difficult to melt the tablets during extrusion, which is disadvantageous from a cost standpoint. Preferably, the pressure during manufacture should be between 0.5 and 3.0 tons/cm^2, which will allow tablets to be obtained that are more durable and still lend themselves well to extrusion.

[0020] The density of the tablets of the present invention is between 0.7 and 1.5.

[0021] From the standpoints of ease of handling and extrusion productivity, it is preferable for the tablets of the present invention to have a diameter (the minor diameter in the case of anisotropy) between 0.1 and 10 mm. The aspect ratio (major diameter/minor diameter) is preferably 2 or less.

[0022]

Embodiments of the Invention

The present invention will now be described in further detail through examples, but the present invention is not limited in any way by these examples.

[0023] Examples 1 to 3

The raw material used in this test was a powder of poly(2,6-dimethyl-1,4-phenylene) ether (hereinafter referred to as PPE) with a density of 0.527 and an intrinsic viscosity (30°C in chloroform) of 0.53 and containing 60 wt% particles with a size of less than 100 μm. The mold consisted of a hollow iron cylinder and an iron piston, with the cylinder measuring 30 mm in outside diameter, 10 mm in inside diameter, and 10 mm in height. The raw material PPE powder and the mold were preheated to 115°C, 0.4 g of the heated PPE powder was packed into the mold, the piston was then fixed, and a pressure of

(1) 1 ton/cm^2,
(2) 2 tons/cm^2, or
(3) 3 tons/cm^2

was applied by a hydraulic compression machine for 10 seconds, after which the molded piece was taken out of the mold. The size and weight of the molded pieces were as follows:

(1) outside diameter: 10.0 mm, height: 5.55 mm, weight: 0.392 g
(2) outside diameter: 10.0 mm, height: 5.34 mm, weight: 0.393 g
(3) outside diameter: 10.0 mm, height: 5.28 mm, weight: 0.392 g

From this we know that the density of the molded pieces were as follows:

(1) 0.899
(2) 0.937
(3) 0.945

No separation or cracking was seen on the surface of the molded pieces. A load was applied to each molded piece, and the load at which the piece broke was measured (hereinafter referred to as the compression strength).

(1) 16.5 kg
(2) 24.7 kg
(3) 30.9 kg

[0024] Example 4

The same operation as in Example 1 was carried out, except that the pressure applied by the hydraulic compressor was changed to 2 tons/cm^2, and the heating temperature of the PPE powder and the mold was changed to 150°C. The size and weight of the obtained molded pieces were as follows:

outside diameter: 10.0 mm, height: 5.02 mm, weight: 0.395 g

From this we know that the density of the molded pieces was 1.002. No separation or cracking was seen on the surface of the molded pieces. The compression strength of the molded pieces was 104.7 kg.

[0025] Example 5

The same PPE powder heated to 150°C, as in Example 4, was used as the raw material, this was press-granulated by a pre-getting roll type of compression molding machine, and PPE tablets were manufactured continuously. The pressure between the rolls was 2 tons/cm^2, and each roll had a hemispherical recess with a diameter of 10 mm. Spherical tablets with a diameter of 10 mm were continuously discharged from the rolls. The tablets thus obtained weighed 0.049 g. From this we know that the density of the molded pieces was 0.993. No separation or cracking was seen on the surface of the molded pieces. The compression strength of the molded pieces was 87.5 kg.

[0026] Comparative Example 1

The same operation as in Example 1 was carried out, except that the PPE powder and the mold were not heated, and were left at room temperature (25°C). The size and weight of the obtained molded pieces were as follows:

outside diameter: 10.0 mm, height: 5.69 mm, weight: 0.393 g

From this we know that the density of the molded pieces was 0.879. The compression strength of the molded pieces was 3.18 kg. Separation and cracking were seen on the surface of the molded pieces.

[0027] Comparative Example 2

The same operation as in Comparative Example 1 was carried out, except that the pressure applied by the hydraulic compressor was changed to 3 tons/cm^2. The size and weight of the obtained molded pieces were as follows.

outside diameter: 10.0 mm, height: 5.57 mm, weight: 0.391 g

From this we know that the density of the molded pieces was 0.894. The compression strength of the molded pieces was 8.65 kg. Separation and cracking were seen on the molded pieces just as in Comparative Example 1.

[0028]

Effect of the Invention

The effect of the method of the present invention is that a polyphenylene ether powder can be easily converted into a shape that is easier to handle and affords greater productivity.

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